

Searching for the Slater Transition in the Pyrochlore $\text{Cd}_2\text{Os}_2\text{O}_7$ with Infrared Spectroscopy

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Infrared reflectance measurements were made on the single crystal pyrochlore $\text{Cd}_2\text{Os}_2\text{O}_7$ in order to examine the transformations of the electronic structure and crystal lattice across the boundary of the metal insulator transition at $T_{MIT} = 226\text{K}$. All predicted IR active phonons are observed in the conductivity over all temperatures and the oscillator strength is found to be temperature independent. These results indicate that charge ordering plays only a minor role in the MIT and that the transition is strictly electronic in nature. The conductivity shows the clear opening of a gap with $2\Delta = 5.2k_B T_{MIT}$. The gap opens continuously, with a temperature dependence similar to that of BCS superconductors, and the gap edge having a distinct $\sigma(\omega) \sim \omega^{1/2}$ dependence. All of these observables support the suggestion of a Slater transition in $\text{Cd}_2\text{Os}_2\text{O}_7$.

Mott's classic paper[1] published over half a century ago has triggered extensive research on correlated electron systems which undergo a metal insulator transition (MIT). Both Mott and Hubbard[2] have suggested that for systems at half filling, the Coulomb repulsion between electrons could split the band, thus producing an insulator. Alternatively, Slater in 1951 suggested that antiferromagnetic order alone could produce an insulator by a doubling of the magnetic unit cell[3]. While there are numerous examples of Slater / spin-density-wave (SDW) insulators in the realm of 1-dimensional (1D) conductors[4, 5], the experimental situation at higher dimensions is less clear. Impossibility of the Slater state in the 2D regime has been recently argued based on dynamical cluster approximation calculations[6]. For 3D solids, spin ordering alone usually generates an energy gap corrupting only a fraction of the Fermi surface so that metallic conductivity persists[7]. In this context the metal-insulator transition in the 3D pyrochlore $\text{Cd}_2\text{Os}_2\text{O}_7$ is exceptionally intriguing since transport and magnetic properties across the MIT boundary appear to be in accord with the Slater mechanism[8]. In this paper we report on the first spectroscopic studies of the MIT in $\text{Cd}_2\text{Os}_2\text{O}_7$. Our analysis of the infrared data reveals that the transition into the insulating state is driven solely by the electronic interactions without significant involvement of the crystal lattice. We discuss new facets of the spin-driven insulating state in a 3D material.

The pyrochlore $\text{Cd}_2\text{Os}_2\text{O}_7$ was first characterized by Sleight et al.[9]. The metal-insulator transition in the resistivity has been found to occur at the same temperature $\simeq 226\text{K}$ as the antiferromagnetic transition in susceptibility measurements. No evidence of concurrent structural changes were detected through X-ray diffraction (XRD) analysis. Thorough examination of transport and magnetism in $\text{Cd}_2\text{Os}_2\text{O}_7$ has been recently reported by Mandrus et al.[8] with a Slater picture delivering a coherent interpretation of all experimental data. This particular mechanism in $\text{Cd}_2\text{Os}_2\text{O}_7$ may be favored by

the fact that Os^{5+} is in the $5d^3$ configuration so that t_{2g} band is near half filling.

The Slater transition is characterized by several hallmarks in the frequency domain which so far remained unexplored in $\text{Cd}_2\text{Os}_2\text{O}_7$. Among them, the specific temperature dependence of the energy gap as well as the frequency dependence of the dissipative response at energies above the gap edge that are distinct from the Mott-Hubbard case[10]. Also the analysis of the IR active phonon modes allows one to verify if a development of charge ordering is concomitant with spin ordering. Despite the fact that IR spectroscopy is ideally suited for directly studying the nature of the MIT in solids, the extremely small size of $\text{Cd}_2\text{Os}_2\text{O}_7$ single crystals so far has rendered these measurements impossible. Spectroscopic tools available in our lab at UCSD are tailored for investigations of microcrystals,[11] allowing us to fill voids in the experimental picture of the insulating state in $\text{Cd}_2\text{Os}_2\text{O}_7$.

The near normal reflectance $R(\omega)$ of $\text{Cd}_2\text{Os}_2\text{O}_7$ was measured from 40 cm^{-1} to 14000 cm^{-1} using a Fourier transform spectrometer and from 12000 cm^{-1} to 35000 cm^{-1} using a grating monochromator. A test involving polarized light displayed no signs of anisotropy, thus unpolarized radiation was used for a detailed study of the temperature dependence. Samples were coated *in situ* with gold or aluminum and spectra measured from the coated surface were used as a reference. This method, discussed previously in detail[12], allows one to reliably obtain the absolute value of the reflectance by minimizing the errors associated with non-specular reflection and small sample size. We inferred the complex conductivity $\sigma_1(\omega) + i\sigma_2(\omega)$ by use of Kramers-Kronig (KK) analysis after extrapolating data to $\omega \rightarrow 0$ and $\omega \rightarrow \infty$. Various low frequency extrapolations (Drude, Hagen-Rubens) were used; however the data did not significantly depend on the particular method of low- ω extrapolation. We employed the usual ω^{-4} dependence for the high-energy extension of the data[13].

FIG. 2: Real part of the optical conductivity as obtained by Kramers-Kronig analysis on a log scale. A gap can be seen to develop continuously as the temperature is reduced. The reduction in spectral weight occurring in the infrared region is compensated by a shift to higher energies. The inset shows the hardening Real part of the optical conductivity as obtained by Kramers-Kronig analysis on a log scale. A gap can be seen to develop continuously as the temperature is reduced. The reduction in spectral weight occurring in the infrared region is compensated by a shift to higher energies. The inset shows the hardening of the 347 cm^{-1} phonon at low T.

tween the two segments at 818 cm^{-1} can be chosen as a quantitative measure of the energy gap 2Δ . The $\omega^{1/2}$ behavior can be recognized at higher temperatures as well. In the latter spectra (Fig.3) the intragap region is best described with a linear dependence. The temperature dependence of the intersection between the $\sigma_1(\omega) \propto \omega$ and $\sigma_1(\omega) \propto \omega^{1/2}$ regions is plotted in the inset of Fig.3.

It is instructive to characterize the development of the energy gap in $\text{Cd}_2\text{Os}_2\text{O}_7$ through the spectra of the effective spectral weight $N_{eff}(\omega) = \frac{120}{\pi} \int_0^\omega \sigma_1(\omega') d\omega'$. The magnitude of $N_{eff}(\omega)$ depicted in Fig.4, is proportional to the number of carriers participating in the optical absorption up to a cutoff frequency ω , and has the dimension of frequency squared. The significant reduction in spectral weight occurring in the intragap region is transferred to the energy region above 3Δ . Interestingly, the spectral weight does not become completely exhausted until 16000 cm^{-1} implying that the energy range as broad as $40\Delta \simeq 104k_B T_{MIT}$ is involved in the metal insulator transition.

Important insights into the origin of the insulating state in $\text{Cd}_2\text{Os}_2\text{O}_7$ may be reached through the analysis of the phonon spectra. The pyrochlore structure belongs to the space group $\text{Fd}\bar{3}\text{m}$ and reveals seven IR active modes[15, 16]. We observe phonon peaks at 86 cm^{-1} , 108 cm^{-1} , 200 cm^{-1} , 347 cm^{-1} , 371 cm^{-1} , 440 cm^{-1} ,

FIG. 4: Effective spectral weight vs. cutoff frequency for temperatures as indicated. The top axis is in energy units normalized by twice the gap energy. The upper inset shows the temperature dependence of the 347 cm^{-1} phonon and the BCS gap function is also plotted, (solid curve). The lower inset shows the temperature dependence of all seven phonons.

the energy gap in the electronic conductivity displayed in Fig.2 which is consistent with the Slater picture of the MIT[19, 20]. The second order transition is in accord with earlier specific heat and magnetic susceptibility data[8].

Further support for the Slater hypothesis in the context of the $\text{Cd}_2\text{Os}_2\text{O}_7$ data is provided by the electronic conductivity. We first note that our data reveals several hallmarks of the Bardeen-Cooper-Schrieffer (BCS) electrodynamics expected for systems with spin density waves[5], including the ratio of $2\Delta/k_B T_{MIT} \simeq 5.1$ (expected in a modified BCS theory that takes into account the scattering of electrons by phonons, as for the canonical SDW Chromium)[21, 22] as well as a characteristic decline of the gap value at non-zero temperatures [19, 20]. The general form of the conductivity spectra is also consistent with the BCS picture where type 2 coherence factors lead to an overshoot between the data at $T \ll T_{MIT}$ and $T > T_{MIT}$ at frequencies above the gap. As pointed

out above, the behavior of the low-T spectra above the gap edge are adequately described with the $\sigma_1(\omega) \sim \omega^{1/2}$ dependence, as is expected for a Slater transition. This finding is important because the $\omega^{1/2}$ dependence is distinct from the $\omega^{3/2}$ dependence observed in the Hubbard limit[10].

Given the experimental evidence discussed above, the Slater mechanism emerges as a viable model of the MIT in $\text{Cd}_2\text{Os}_2\text{O}_7$. Therefore, this compound may be the first well documented case of a three-dimensional SDW material[23], subject to further direct verification of the spin structure. An unexpected feature of the antiferromagnetically-driven MIT is a mismatch between the $T_{MIT} \simeq 200\text{K}$ and the frequency range involved in the redistribution of the spectral weight in the insulating state $\Omega \simeq 20000\text{K}$. Similar mismatch is commonly found throughout the spectroscopic studies of the so-called pseudogap state in high- T_c superconductors[24],

in which antiferromagnetic fluctuations are perceived as a likely cause of the pseudogap state. Finally, it is worth mentioning that other pyrochlore compounds reveal very different properties at the verge of the metal-insulator transition. For instance, the transition to the insulating regime in the closely related $\text{Tl}_2\text{Ru}_2\text{O}_7$ system appears to be of first order and additionally is accompanied by charge ordering effects judging from the transformations of the phonon spectra[25]. It is yet to be determined what microscopic factors define the peculiar character of the MIT in $\text{Cd}_2\text{Os}_2\text{O}_7$.

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